

Comment on "Generalized Curie-Weiss law"

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Recently, Fähnle and Souletie [J. Phys. C 17, L469 (1984)], as well as Arrott [Phys. Rev. B 31, 2851 (1985)], have demonstrated that the susceptibility of some spin models is very well approximated by a power law, not only in the critical regime, but also over the whole paramagnetic temperature range. The results may be described by a generalized Curie-Weiss law with two adjustable parameters. To fix the values of these parameters the authors have used different methods, which are discussed and compared in this Comment.

In 1983 Souletie and Tholence<sup>1</sup> pointed out that the temperature dependence of the paramagnetic susceptibility  $\chi$  of crystalline nickel may be very well represented by a power law over the whole temperature range above the Curie temperature  $T_c$ , up to  $T = 3T_c$ . This was a great surprise, because usually it is assumed that a power law is only valid in the small critical regime very close to  $T_c$ , whereas the temperature dependence outside the universality range is expected to be very complicated. Fähnle and Souletie<sup>2</sup> and, independently, Arrott<sup>3</sup> have shown that the Padé approximants and the high-temperature series expansions, respectively, for the susceptibility of some localized spin models indeed are very well approximated by a power law over the whole temperature range. Their results may be summarized by a formula called the generalized Curie-Weiss law by Arrott, which is most generally written in the form

$$\chi T/T_0 = (1 - T^*/T)^{-\gamma^*}, \tag{1}$$

where  $T_0$  is Curie's constant. It is essential for this generalized Curie-Weiss law that a nonlinear<sup>1,2</sup> temperature variable  $(T - T^*)/T$  be used instead of the common linear variable  $(T - T_c)/T_c$ . The purpose of this paper is to distinguish the different methods applied by the authors to fix the parameters  $T^*$  and  $\gamma^*$  of the generalized Curie-Weiss law.

In the paper of Fähnle and Souletie<sup>2</sup> the authors demand that the very-high-temperature expansion of Eq. (1),

$$\chi T/T_0 = (1 + \gamma^* T^*/T), \tag{2}$$

be equivalent to the high-temperature-series expansion truncated at the term linear in  $1/T$

$$\chi T/T_0 = [1 + a_1(J/k_B T)] \tag{3}$$

Here  $J$  and  $k_B$  are the nearest-neighbor exchange integral and Boltzmann's constant, and  $a_1$  is the "first" coefficient in the series expansion corresponding to the power  $(1/T)^1$ . [There is a difference in notation between Refs. 2 and 3: The quantity corresponding to  $\gamma^*$  is denoted by  $\hat{\gamma}(\infty)$  in Ref. 2, whereas the symbol  $\gamma^*$  is reserved for the effective Kouvel-Fisher exponent  $\gamma^* = (T - T_c)\chi d\chi^{-1}/dT$ .] Alternatively, the same procedure may be performed with the expansion of the Padé approximants, yielding Eq. (3) with

$$a_1 = \gamma_c(p_1 - q_1) \tag{4}$$

for Ising models and

$$a_1 = (p_1 - q_1) + \gamma_c \frac{k_B T_c}{J} \tag{5}$$

for Heisenberg models, where  $p_1$  and  $q_1$  are the "first" coefficients in the numerator and denominator of the approximants used in Ref. 2, and  $\gamma_c$  is the critical exponent describing the temperature dependence of  $\chi$  for  $T \rightarrow T_c$ , estimated from the Padé approximants. It should be noted that for the Heisenberg systems the values for  $a_1$  determined from Eq. (5) are not exactly the same as those for the coefficients  $a_1$  of the series expansion. This results from the uncertainties in the values of  $\gamma_c$ ; indeed, the  $\gamma_c$  estimates from the Padé approximants listed in Tables I and II are not universal for the three-dimensional Heisenberg models.

Comparison of Eqs. (2) and (3) yields

$$\gamma^* \frac{k_B T^*}{J} = a_1 \tag{6}$$

Choosing an ansatz for one parameter, say  $T^*$ , Eq. (6) fixes the second parameter  $\gamma^*$ , and for each choice the very-high-temperature expansion, Eq. (2), is equivalent to Eq. (3).

The question then remains whether the so-obtained generalized Curie-Weiss law fits the susceptibility data in a large temperature range or only at high temperatures. In the paper of Fähnle and Souletie<sup>2</sup> it has been shown that the choice  $T^* = T_c$  (with  $T_c$  determined from the Padé approximants) provides a reasonably good fit, for some systems even an excellent fit from  $T_c$  up to infinitely high temperatures, e.g., for the bcc and fcc Ising model, the  $S = 1$  bcc and fcc Heisenberg model, and the  $S = \infty$  simple cubic (sc) Heisenberg model (Figs. 1 and 2 of Ref. 2). This means that in the latter systems the high-temperature exponent  $\gamma^* = a_1(J/k_B T_c)$  [Eq. (14) of Ref. 2] obtained from Eq. (6) with  $T^* = T_c$  is very close (Tables I and II) to the critical exponent  $\gamma_c$ , a result which is consistent with the original conclusions of Souletie and Tholence<sup>1</sup> on the susceptibility of crystalline nickel. The generalized Curie-Weiss law then reads

$$\chi T/T_0 = (1 - T_c/T)^{-\gamma_c}, \tag{7}$$

TABLE I.  $T^*$  and  $\gamma^*$  obtained by different methods. The values for  $T_c$  and  $\gamma_c$  are from Ref. 5. Note that for both the  $S = \frac{1}{2}$  and the  $S = \infty$  Heisenberg model the exchange integral  $J$  is defined by the same Hamiltonian  $H = -(J/S^2) \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ , whereas in the paper of Arrott,  $J$  is defined by  $H = -2J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$  for  $S = \frac{1}{2}$  and by  $H = -(2J/S^2) \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$  for  $S = \infty$ .

|                   |     | $\frac{J}{k_B T^*}$  |             |                      | $\gamma^*$                     |                                    | $\gamma_c$ | $\gamma^* = \gamma_p$ |
|-------------------|-----|----------------------|-------------|----------------------|--------------------------------|------------------------------------|------------|-----------------------|
|                   |     | Best fit<br>(Ref. 3) | $T^* = T_c$ | $T^* = T_p/\gamma_p$ | Best fit<br>(Ref. 3)           | $\gamma^* = a_1 \frac{J}{k_B T_c}$ |            |                       |
|                   |     |                      |             |                      | $a_1$ from<br>series expansion | $a_1$ from<br>Eqs. (4) and (5)     |            |                       |
| $S = \frac{1}{2}$ | sc  | 0.3275               | 0.296       | 0.5                  | 1.89                           | 1.776                              | 1.42       | 3.0                   |
|                   | bcc | 0.2074               | 0.1974      | 0.25                 | 1.63                           | 1.5792                             | 1.39       | 2.0                   |
|                   | fcc | 0.1266               | 0.1241      | 0.125                | 1.52                           | 1.4892                             | 1.43       | 1.5                   |
| $S = \infty$      | sc  | 0.694                | 0.6916      | 0.75                 | 1.377                          | 1.3832                             | 1.38       | 1.5                   |
|                   | bcc | 0.4804               | 0.48635     | 0.5                  | 1.2766                         | 1.297                              | 1.37       | 1.3333                |
|                   | fcc | 0.3084               | 0.3147      | 0.3                  | 1.242                          | 1.2588                             | 1.38       | 1.2                   |

and Eq. (2) yields the high-temperature behavior

$$\chi^{-1} \sim (T - T_c^{MF}), \quad (8)$$

with a nontrivial prediction  $T_c^{MF} = \gamma_c T_c$ , which can easily be checked experimentally.

For other systems (e.g., the  $S = \frac{1}{2}$  sc Heisenberg model) the exponent  $\gamma^* = a_1(J/k_B T_c)$  is different from  $\gamma_c$ . Equation (7) then is accurately valid only at temperatures not too far from  $T_c$ , and should be replaced by

$$\chi T/T_0 = (1 - T_c/T)^{-\gamma^*} \quad (9)$$

at high temperatures. When inserting the correct value of  $T_c$  as obtained, for example, from the Padé approximants as well as  $\gamma^* = a_1(J/k_B T_c)$ , Eq. (9) yields an asymptotically correct relation for the very-high-temperature susceptibility  $\chi^{-1} \sim (T - T_c^{MF})$  with  $T_c^{MF} = \gamma^* T_c$ . Obviously, there is in all cases, for finite coordination number  $q$ , a well-defined  $\gamma^* \neq 1$ , the presence of which is reflected by the fact that  $T_c^{MF} = \gamma^* T_c$  instead of  $T_c^{MF} = T_c$ .

The changeover from the critical exponent  $\gamma_c$  to the exponent  $\gamma^*$  may be conceived as follows: In the critical regime the thermal correlation length  $\xi(T)$  is much larger than the lattice constant. Therefore, we observe a universal

critical exponent  $\gamma_c$ , independent of the interaction details (lattice structure, spin quantum number). For increasing  $T$  (decreasing  $\xi$ ) things become sensitive to the structure of the lattice, and, therefore, the high-temperature exponent  $\gamma^*$  in general depends on the spin quantum number  $S$ , the lattice structure, and the interaction range. Thereby, there are two general trends in the  $\gamma^*$  data (Fig. 3 of Ref. 2 and Tables I and II): For small coordination number (e.g.,  $q = 6$ , sc lattice)  $\gamma^*$  is larger than  $\gamma_c$ , the value of  $\gamma^*$  decreases with increasing  $q$  and for  $q \rightarrow \infty$  molecular-field theory holds with  $\gamma^* = 1$ . Furthermore, the values of  $\gamma^*$  are larger than  $\gamma_c$  for all  $S = \frac{1}{2}$  Heisenberg models and decrease with increasing  $S$ . The agreement between  $\gamma_c$  and  $\gamma^*$ , therefore, is best for medium values of  $q$  and  $S$ , e.g., for the  $S = 1$  bcc ( $q = 8$ ) Heisenberg model.

In the paper of Arrott<sup>3</sup> the author is proceeding on two slightly different lines. In a first method, Arrott conceives the quantities  $T^*$  and  $\gamma^*$  as adjustable parameters for an optimum fit to the susceptibility data of the high-temperature series in the whole temperature range. The result of his analysis is  $T^* \simeq T_c$ , in agreement with our choice for  $T^*$ , and for the best-fit values of  $\gamma^*$  he obtains figures close to our values  $\gamma^* = a_1(J/k_B T_c)$  (see Table I). Alternatively, Arrott proposes a method to obtain rough estimates for the

TABLE II.  $T^*$  and  $\gamma^*$  obtained from  $T^* = T_c$  and  $\gamma^* = a_1(J/k_B T_c)$  [Eq. (6)]. The values for  $T_c$  and  $a_1$  are from Ref. 5 for the Heisenberg models and from Ref. 6 for the Ising models.

|                                    | Ising models |             |           | Heisenberg models |        |          |                   |          |          |                   |        |          |
|------------------------------------|--------------|-------------|-----------|-------------------|--------|----------|-------------------|----------|----------|-------------------|--------|----------|
|                                    | sc           | bcc         | fcc       | $S = 1$           |        |          | $S = \frac{3}{2}$ |          |          | $S = \frac{5}{2}$ |        |          |
|                                    | sc           | bcc         | fcc       | sc                | bcc    | fcc      | sc                | bcc      | fcc      | sc                | bcc    | fcc      |
| $\frac{J}{k_B T_c}$                | 0.218 156    | 0.156 178 9 | 0.101 767 | 0.383             | 0.2632 | 0.167 95 | 0.4385            | 0.304 74 | 0.195 58 | 0.506             | 0.3543 | 0.228 15 |
| $\gamma^* = a_1 \frac{J}{k_B T_c}$ | 1.309        | 1.2494      | 1.2212    | 1.532             | 1.404  | 1.344    | 1.462             | 1.354    | 1.304    | 1.417             | 1.323  | 1.278    |
| ( $a_1$ from series expansion)     |              |             |           |                   |        |          |                   |          |          |                   |        |          |
| $\gamma^* = a_1 \frac{J}{k_B T_c}$ | 1.309        | 1.2494      | 1.2212    | 1.547             | 1.414  | 1.359    | 1.467             | 1.359    | 1.304    | 1.407             | 1.308  | 1.268    |
| [ $a_1$ from Eqs. (4) and (5)]     |              |             |           |                   |        |          |                   |          |          |                   |        |          |
| $\gamma_c$                         | 1.25         | 1.25        | 1.25      | 1.39              | 1.385  | 1.39     | 1.38              | 1.38     | 1.375    | 1.365             | 1.36   | 1.365    |

quantities  $T^*$  and  $\gamma^*$ . The quantity  $T^*$  is approximated by

$$T^* = T_p / \gamma_p, \quad (10)$$

where  $T_p$  is the molecular-field result for the paramagnetic Curie temperature,

$$T_p = a_1 \frac{J}{k_B}. \quad (11)$$

The exponent  $\gamma_p$  is determined from a comparison of the expansion coefficients of Eq. (1), with  $\gamma^* = \gamma_p$  and the expansion coefficients of the high-temperature series, respectively, both truncated at the term  $(1/T)^2$ . This yields

$$1/\gamma_p = 2a_2/a_1^2 - 1. \quad (12)$$

Of course Eq. (6) is again fulfilled for  $\gamma^* = \gamma_p$  and  $T^* = T_p/\gamma_p$ , with  $T_p$  given by Eq. (11), and thus the asymptotic temperature dependence of  $\chi$  is described correctly. However, the fit to the susceptibility data in the whole temperature range is considerably improved when replacing  $\gamma_p$  and  $T_p/\gamma_p$  by the best-fit values obtained by Arrott or by the values  $\gamma^* = a_1(J/k_B T_c)$  and  $T^* = T_c$  (with the correct figures for  $T_c$ ) used by Föhnle and Souletie. Nevertheless, the exponent  $\gamma_p$  is a useful quantity because it exhibits the same trends in terms of spin quantum number and lattice

structure as the best-fit values and those calculated from  $\gamma^* = a_1(J/k_B T_c)$ . As an advantage,  $\gamma_p$  and  $T_p$  are much easier to determine than the quantities  $\gamma^* = a_1(J/k_B T_c)$  and  $T_c$ , especially for systems for which only a small number of coefficients in the high-temperature expansion are known, so that the determination of  $\gamma_c$  and  $T_c$  is uncertain.

In Table I we compare the results for  $T^*$  and  $\gamma^*$  for the  $S = \frac{1}{2}$  and the  $S = \infty$  Heisenberg models calculated by the different methods. It is obvious that the best-fit values obtained by Arrott are very close to those used by Föhnle and Souletie. Table II exhibits the parameters determined by the latter authors for those models not considered by Arrott. Especially interesting is the  $S = 1$  bcc Heisenberg model, because in this case the high-temperature exponent is very close to the critical exponent (see above), and, furthermore, the model might be appropriate for the discussion of  $\alpha$ -iron. For the two-dimensional Ising models the values for  $z_c = \exp(-2J/k_B T_c)$  are known exactly,<sup>4</sup> namely,  $z_c = 2 - \sqrt{3}$ ,  $\sqrt{2} - 1$ , and  $1/\sqrt{3}$  for the honeycomb ( $q = 3$ ), quadratic ( $q = 4$ ), and triangular ( $q = 6$ ) lattices, respectively. Because the coefficient  $a_1$  is given by the coordination number  $q$  in these cases, the exponent  $\gamma^* = a_1(J/k_B T_c)$  may be calculated exactly, yielding  $\gamma^* = 1.9754$ ,  $1.7627$ , and  $1.6479$ , whereas the critical exponent  $\gamma_c$  is given by  $\gamma_c = 1.75$  for the two-dimensional Ising models.

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